Dendrite Growth of Aluminum-Copper Alloy Reinforced with Continuous Alumina Fibers

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Directional solidification studies were carried out in continuous alumina fiber reinforced Al–4.5 and 15 mass%Cu alloy composites in order to clarify the influences of fibers on crystal growth of matrix alloy in the composites. The specimens were designed to have an inner composite region and an outer unreinforced bulk region in order to characterize the dendrite morphology in both regions under the same conditions. The composite specimen was produced by a pressure infiltration process, subsequently remelted, and then directionally solidified. In the composite region of the specimens, the shape of the dendrite was distorted by the presence of fibers, with the primary dendrite tip position being located about 450–750 μm below that in the bulk region. The difference of undercooling was estimated to be 1.2–1.7 K. The concentration of copper at the dendrite tips was 0.05–0.15 mass% higher than that in the bulk region, and the tip composition increased as the fiber interstices became smaller. The solutal undercooling was estimated to be 0.9–2.7 K, so that solutal diffusion field around dendrite tip could have governed the dendrite tip undercooling. Furthermore, in the composite region, the primary dendrite arm spacing decreased to about 70% of that in the bulk region on an average. A model based on the continuity of liquid phase among fibers reveals how fibers influence both the concentration of copper on the dendrite tips and the size of the lateral solute diffusion field.

(Received September 11, 2000; Accepted December 1, 2000)

Keywords: composites, directional solidification, continuous fiber, solute diffusion, dendrite, alumina ceramics

1. Introduction

An advantage of liquid state fabrication for producing metal matrix composites (MMCs) might be the expandability of freedom of phase selection and the fabrication of complicated components at a lower cost. However, the solidification microstructure of the matrix around reinforcements becomes quite different from that of unreinforced alloy. This because the crystal growth of the primary and eutectic phases in composites is led by thermal, solutal and geometrical influences of the fibers, especially in regions where the fiber spacing is smaller than the primary and secondary arm spacings. The microstructural changes caused by the restriction of solute diffusion and the wettability between reinforcement and matrix alloy have been reported in metallic system specimens with continuous fibers, and in organic system specimens. Few papers, however, have broached dendrite growth morphology that could occur in the fabrication of commercial MMCs, and the solidification mechanism of the matrix alloy among fibers is still unrevealed.

The purpose of this work is to evaluate the thermal, solutal and geometrical influences of fibers on the dendrite growth of metal matrix composites. The study was done by analyzing dendrite tip undercooling and primary dendrite arm spacing, and by comparing experimental data with an analytical model based on the continuity and solutal diffusion in the liquid phase.

2. Experimental Procedure

Matrix alloys of Al–4.5 and 15 mass%Cu (abbreviate to %) were prepared from 99.99%Al and 99.99%Cu, and continuous alumina fiber (ALTEX, γ-Al2O3, diameter = 15 μm: Sumitomo Chemical Co., Ltd.) was used for reinforcement. Composites and unreinforced specimens were fabricated by the axial pressure infiltration process. The matrix alloy and binder free fiber preform were placed between a piston and a cylinder mold made of graphite, and then preheated to a temperature of 1073 K in a vacuum atmosphere of 2–3 Pa. The piston was then pressed down at a pressure of 14 MPa to infiltrate the molten metal into the fiber preform. The specimen consisted of an inner composite region and an outer unreinforced bulk region; this as molten alloy initially infiltrated into the gap between the fibers and mold, relatively a larger space, then pushed the fibers to the center laterally during the pressure infiltration process.

Directional solidification study was carried out using a Bridgeman type furnace with a water-cooled quenching unit. The composite specimen (5 mm diameter and 35 mm length, as shown in Fig. 1) was placed in a graphite crucible, pre-

![Fig. 1. Crucible and composite specimen for the directional solidification experiment.](image-url)
Table 1  Experimental conditions (C0: alloy concentration, G: temperature gradient, R: growth velocity, Vc: cooling velocity).

<table>
<thead>
<tr>
<th>No</th>
<th>C0 [mass%]</th>
<th>G [K/mm]</th>
<th>R [mm/s]</th>
<th>Vc [×10^{-3} K/s]</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>4.5</td>
<td>2.6</td>
<td>2.2</td>
<td>2.9</td>
</tr>
<tr>
<td>2</td>
<td>14.8</td>
<td>2.3</td>
<td>39.0</td>
<td>71.0</td>
</tr>
</tbody>
</table>

heated at a steady 1053 K for 1 hour, and then directionally solidified at a velocity of 2–40 μm/s and a temperature gradient of 2.5 K/mm in an argon atmosphere (Table 1). Half of the original specimen was solidified, and then quenched with a water spray. The longitudinal (in parallel with growth direction and fiber array) and transverse microstructures in both the composite and bulk regions were examined by a standard optical microscope. The morphology of the dendrite and primary dendrite arm spacings were investigated, and the composition of copper at the dendrite tips was measured on several transverse sections by an electron probe micro analyzer (EPMA).

3. Results and Discussion

3.1 Microstructure observation

Figure 2 shows a typical micrograph at the longitudinal cross section of the alumina/Al–15%Cu alloy composite specimens. The continuous alumina fibers (dark lines) are shown as short fibers because of the difference of alignment between the fibers and the cutting surface. As shown on the left side in Fig. 2(a), the trunk and the secondary arms of the dendrites are clearly observed in the bulk region. The dendrites, however, are unclear and hardly recognizable in the composite region as shown on the right side of the figure; so the contours of the dendrite shape were illustrated in Fig. 2(b) by tracing an edge of α phase in Fig. 2(a). The dendrites in the composite region are deformed and the secondary arms are shortened by the presence of fibers.

In order to comprehend an accurate dendrite morphology in the composite region, the composition of the copper in the dendrite was measured by EPMA. Figure 3 shows a back scattered electron image and the composition mapping of copper in the alumina/Al–15%Cu specimen. The dendrite seems to be deformed as shown in Fig. 3(a). The solid line in Fig. 3(b) shows the composition of 2.65% Cu, which is maximum in solid at 881 K according to the equilibrium phase diagram. Since the sample was quenched at 881 K, a solid line can indicate the solid/liquid interface. Therefore, the true shape of the dendrite tip is elliptic or parabolic at quenching. Similar dendrite shapes are observed in Al–4.5%Cu specimens. It is presumed that the development of the secondary arm was restrained and the dendrite shape was changed to a cell-like one in the limited space where the dendrite is completely surrounded by fibers as shown in Fig. 3.

3.2 Dendrite tip undercooling

The positions of dendrite tips in the composite region (marked ‘B’) are located below those in the bulk region (marked ‘A’) as shown in Fig. 2. From the investigation of several transverse sections in the quenched mushy zone at an interval of 20–30 μm, it was realized that the differences between dendrite tip positions in the composite and bulk regions were at least 750 μm in Al–15%Cu specimens solidified at 39 μm/s, and 450 μm in Al–4.5%Cu specimens solidified at 2.2 μm/s. These distances corresponded to undercoolings of 1.7 K and 1.2 K for Al–15%Cu and Al–4.5%Cu specimens.

Fig. 3  Longitudinal section through the dendrite tips in the composite region of alumina/Al–15 mass%Cu alloy composites. (a) Back-scattered electron image. (b) Copper composition map with EPMA analysis. The solid line shows 2.65 mass%Cu.

Fig. 2  Longitudinal section through the quenched interface in alumina/Al–15%Cu alloy composites. (a) Micrograph. (b) Schematic illustration of the primary α phase.
respectively. The difference in undercooling could be explained based on an idea that tip undercooling is influenced by fibers. Generally, the undercooling of a dendrite tip is described as follows,

$$\Delta T = \Delta T_c + \Delta T_R + \Delta T_k$$  \hspace{1cm} (1)

where, $\Delta T_R$ is a function of the dendrite tip radius $r$ described as,

$$\Delta T_R = \frac{\Gamma}{r}, \quad \Gamma = \frac{\sigma}{\Delta S_f}$$  \hspace{1cm} (2)

$\Gamma$ is the Gibbs-Thomson coefficient [Km], $\sigma$ is the interfacial energy between solid and liquid [J/m²], and $\Delta S_f$ is the entropy of fusion [J/mol K]. If the tip radius is changed because of contact with fibers or by the increases in the solute content around dendrite tip, $\Delta T_R$ can be changed. However, the dendrites untouched by fibers, the difference in tip radius between the composite and the bulk regions cannot be detected. Dendrite tip radii are 7 µm and 30 µm for Al–15%Cu and Al–4.5%Cu specimens, respectively. Supposing $\Gamma = 2.4 \times 10^{-7}$ [Km] then $\Delta T_R$ is estimated at 0.068 K and 0.016 K for 7 µm and 30 µm of tip radius, respectively. Therefore, the contribution of $\Delta T_R$ could be negligible. $\Delta T_k$ is described as,

$$\Delta T_k = \frac{1}{\mu_0} V, \quad \mu_0 = \frac{\Delta H_f}{R T_M^2}$$  \hspace{1cm} (3)

where, $V$, $\Delta H_f$, $R$, $T_M$ and $V_0$ are solidification velocity [m/s], enthalpy of fusion [J/mol], gas constant, melting point [K], and velocity of sound [m/s], respectively. Since the $\alpha$ phase in an Al–Cu system has a nonfacet growth, $\Delta T_k$ is estimated at less than $10^{-5}$–$10^{-6}$ K from the physical parameter presented in Ref. 9), and finally can be neglected.

Composition analysis was performed to evaluate the influence of the composition on tip undercooling. Since each dendrite tip scattered due to the fiber distribution around dendrites, specimens were sliced at intervals of 20–30 µm, and the locations of dendrite tips in each transverse section were investigated. Figure 4 shows the relationship between tip compositions and the positions of dendrite tips in the composite region. The positions are standardized by the positions of dendrite tips in the bulk region. The solid lines in the figure are the calculated compositions from an equilibrium phase diagram. In the composite region, the tip compositions are higher than those in the bulk region, and increase with increasing differences in dendrite tip position between the composite and the bulk regions.

### 3.3 Lateral growth of $\alpha$ phase

Since it seems that the difference in dendrite tip position between the composite and the bulk regions becomes larger as the fiber interstices become smaller, we defined the mean fiber interstices and evaluated the relationship to the tip composition as follows:

$$d_l = \frac{d_{max} + d_{min}}{2}$$  \hspace{1cm} (4)

where, $d_{max}$ and $d_{min}$ are the length [m] of the major and minor axes of an ellipse region that was assumed from the primary $\alpha$ phase in Fig. 5. Figure 6 shows the relationship between tip composition and parameter $d_l$. The tip composition increases with the decreasing value of $d_l$. Since solutal diffusion and dendrite growth could be influenced by the fiber configuration, we also defined the local mean fiber volume fraction as shown in Fig. 5, and evaluated the relationship to the tip composition as:

$$V_d = d_1 + d_2 + d_3 + \cdots + d_n$$  \hspace{1cm} (5)

where, $n$ is the number of fibers surrounding the $\alpha$ phase, $d_n$ is the nearest fiber distance around the $\alpha$ phase, $r_f$ is the radius of fibers. As shown in Fig. 7, it seems that the tip composition also tends to increase when the value of $V_d$ increases. According to Figs. 6 and 7, the value of $d_l$ in eq. (4) leads to a better agreement on the tip composition compared with the value of $V_d$ in eq. (5).

Since the dendrite arm spacing is hard to recognize due to the coarsened secondary dendrite arm in the composite region, the primary $\alpha$ phase distance is used instead of the dendrite arm spacing parameter. Figure 8 shows the primary dendrite arm spacing ($\lambda_1$) or the primary $\alpha$ phase distance ($\lambda_C$) as functions of the thermal gradient and growth velocity. The relationship in the bulk can be expressed as:

$$\lambda_1 = a (C_0^{1/4} \cdot R^{-1/4} \cdot G^{-1/2})$$  \hspace{1cm} (6)

where $a$ is a constant ($a = 263$) for the bulk, following the relationship suggested by Kuru and Fisher, Hunt and
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Fig. 6 Relationship between the concentration of copper at the primary α tip and the sizes of fiber interstices for alumina/Al-4.5 mass% Cu alloy composites.

Fig. 7 Relationship between the concentration of copper at the primary α tip and the local mean fiber volume fraction.

Fig. 8 Solidification velocity and temperature gradient dependence of primary dendrite arm spacing ($\lambda_i$) and primary α phase distance ($\lambda_C$).

Trivedi,12,13) In the composite region, the primary α phase distance ($\lambda_C$) is scattered from 300 μm to 620 μm in fixed conditions. The average of $\lambda_C$ is 460 μm and is almost 70% smaller than the value of $\lambda_1$ in the bulk region (660 μm in the same condition). The geometrical distribution of continuous fibers could influence the dendrite growth because of the changes in the solutal diffusive condition.

The diffusion layer around the dendrite tip is examined by the analytical model shown in Fig. 9. The difference ($\Delta C_{LB}$) between the initial composition ($C_0$) and the composition ($C_L$) at a particular radius ($r$) is given by:

$$\Delta C_{LB} = C_L - C_0 = \Delta C^*_L(r - r_B)/(r_B - r)$$  \hspace{1cm} (7)

$$\Delta C^*_L = C^*_L - C_0$$  \hspace{1cm} (8)

where, $r_B$ is the size of diffusion layer, $r_i$ is a half of the trunk of the primary dendrite arm, $C^*_L$ is the composition in liquid phase at the solid/liquid interface.5) The amount of excess Cu contents ($\Delta M_B$ (higher than $C_0$)) in bulk region are given by eq. (9) and eq. (10) under the assumption of a constant gradient of the composition profile:

(a) Hemispherical diffusion field

$$\Delta M_B = \int_{r_i}^{r_B} \Delta C_{LB} \cdot 2\pi r^2 dr$$

$$= \pi \Delta C^*_L (r_B - r_i)(r_B^3 + 2r_B r_i + 3r_i^2)/6$$  \hspace{1cm} (9)

(b) Cylindrical diffusion field

$$\Delta M_B = \int_{r_i}^{r_B} \Delta C_{LB} \cdot 2\pi r dr$$

$$= \pi \Delta C^*_L (r_B - r_i)(r_B + 2r_i)/3$$  \hspace{1cm} (10)

In order to express the diffusion field among the fibers, an effective diffusion coefficient in eq. (11) is estimated by comparing the two dimensional solute diffusion simulation and the one dimensional analytical calculation shown in eq. (12).

$$\frac{\partial C}{\partial t} = D_{\text{comp}} \left( \frac{\partial^2 C}{\partial x^2} + \frac{\partial^2 C}{\partial y^2} \right)$$  \hspace{1cm} (11)

$$C = 1 - \frac{4}{\pi} \sum_{n=0}^{\infty} \frac{(-1)^n}{(2n+1)} \exp \left( \frac{D_{\text{comp}}(2n+1)^2\pi^2 t}{4} \right)$$  \hspace{1cm} (12)
\[ \times \cos \left( \frac{(2n - 1)\pi(1 - x)}{2} \right) \]  

(12)

where, \( dt \) is the infinite time and \( D_{\text{comp}} \) is the effective diffusion coefficient of Cu in molten Al alloy. The effective diffusion coefficient of solute among the fibers becomes smaller than that in the bulk region. So that, the diffusion length \( r_c \) could be smaller than \( r_B \), and the slope of Cu distribution \( (C_{C}^{+} - C_{0})/(r_C - r_f) \) could be larger than the case in the bulk region. In the same way as in the bulk region, as described in eq. (9) and eq. (10), the amount of excess Cu content (\( \Delta M_C \)) in the composite region is given by:

(a) Hemispherical diffusion field

\[ \Delta M_C = \int_{r_f}^{r_C} \Delta C_{LC} \cdot 2\pi r^2 dr \]

\[ = \pi \Delta C_{L}^{*} (r_C - r_f) (r_C^2 + 2r_C r_f + 3r_f^2)/6 \]  

(13)

(b) Cylindrical diffusion field

\[ \Delta M_C = \int_{r_f}^{r_C} \Delta C_{LC} \cdot 2\pi r dr \]

\[ = \pi \Delta C_{L}^{*} (r_C - r_f) (r_C + 2r_f)/3 \]  

(14)

From eqs. (9), (10), (13) and (14), the relationship between diffusion size and volume fraction are given as in eqs. (15) and (16), under the assumptions that; (i) the whole amount of excess Cu content in the diffusion field of the bulk region is equal to that in the composite region: \( \Delta M_B = \Delta M_C \), (ii) \( r_f \) is almost zero because of \( r_f \ll r_B \) and \( r_f \ll r_C \), and (iii) the amount of Cu content which diffuses per second in the bulk region is equal to that in the composite region at steady state growth.

(a) Hemispherical diffusion field

\[ \frac{r_C}{r_B} = \left( \frac{D_{\text{comp}}}{D_{\text{bulk}}} \right)^{1/4} \]  

(15)

(b) Cylindrical diffusion field

\[ \frac{r_C}{r_B} = \left( \frac{D_{\text{comp}}}{D_{\text{bulk}}} \right)^{1/3} \]  

(16)

The relationship between \( r_C/r_B \) and \( V_f \) is summarized in Fig. 10, and the value of \( r_C/r_B \) decreases with increasing \( V_f \). Furthermore, the ratio of excess Cu content at dendrite tips between the bulk and composite regions \( (\Delta C_{B}^{+}/\Delta C_{C}^{+} \) is calculated as 1.9 when the dendrite arm spacing decreases to 70% in the bulk region \( (r_c/r_B = 0.7) \). On the contrary, values of 1.8–3.1 of \( \Delta C_{B}^{+}/\Delta C_{C}^{+} \) are experimentally detected by Fig. 4. Therefore, the fiber constrains the solute diffusion in the transverse direction, and the lower diffusivity could increase the Cu content at the dendrite tip. The pile up of copper solute among the fibers could increase undercooling of dendrite tips and delay dendrite growth.

4. Conclusion

The influences of fibers on dendrite growth of a metal matrix are investigated in directional solidification studies for continuous alumina fibers/Al–4.5 and 15 mass%Cu composite alloys. The results obtained are summarized as follows:

1) The dendrite morphology is significantly influenced by the fiber contribution: the secondary dendrite arm disappears and the dendrite shape changes to become cell-like in the small region where the dendrite is completely surrounded by fibers.

2) The undercoolings of the dendrite tips in the composite region are 1.2–1.7 K larger than that in the bulk region. The undercooling increases with the increasing of the distance between the dendrite tip position in the composite region and that in the bulk region, and with a decreasing of the lateral solidification area surrounded by fibers.

3) The primary (α) phase distance \( (\lambda_C) \) in the composite region decreases to 70% of the primary dendrite arm spacing \( (\lambda) \) in the bulk region. This is explicable using a model that evaluates the influence of the fibers on solute diffusion at the dendrite tips. Furthermore, the results of the model indicates that the solute, which piles up in the transverse direction, could increase the solute content in front of the dendrite tip.

Acknowledgment

The present study was partially supported by the New Energy and Industrial Technology Development Organization (NEDO).

REFERENCES